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A Process for Composite Fabrication by Silicon Slurry Coating of Continuous Fiber Tows

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SUMMARY

A process is described for coating continuous small-diameter fiber tows with silicon-filled polymer slurries. This can enable fabrication of complex shaped ceramic matrix composites such as those based on reaction-bonded-silicon-nitride matrices. The coating process sequence includes sizing removal, drying, spreading, dip-coating, drying, and die consolidation to infiltrate a silicon-filled polymer slurry into interstices of the fiber tows. Sizing removal from the as-received fibers was accomplished by using an ultrasonic cleaner with water-isopropyl alcohol solutions at elevated temperatures (30 to 60 °C). Measurements of surface tension, viscosity, and density of various slurry concentrations are listed. The coating thickness applied on the fiber surface can be correlated with the modified small wire theory. Optical microscopy of cross-sections of coated samples showed the degree of infiltration achieved. The overall diameters of the coated tows depended primarily on the drying (die) temperatures used in the drying furnace, which in turn appears related to the vapor pressure of the solvent evaporating at those temperatures. Finally, the effect of the coating process on the tensile strength of Nicalon fiber is also examined.

INTRODUCTION

SiC fiber-reinforced reaction-bonded-silicon-nitride (SiC/RBSN) composite materials are being studied as possible structural materials for high temperature applications (refs. 1 to 3). Recent research efforts are aimed at improving oxidation resistance and first matrix cracking strength, providing graceful failure (multiple cracking) for random overloads at both room and elevated temperatures, and developing a capability for low cost and near net shape fabrication. In general, there are two kinds of continuous SiC fibers that are used for reinforcement in RBSN materials. The first kind is the multi-filament continuous tow derived from polymeric precursors. This is typically represented by the small-diameter (<20 μm) SiC Nicalon fiber from Nippon Carbon (ref. 4). Recent studies (refs. 5 and 6) have shown that the reduction of oxygen content to <1 wt% of the fiber's composition improves the thermal stability and the high temperature properties of these polymer-derived fibers. The other kind of continuous SiC fiber is produced as monofilament by chemical vapor deposition. This includes the Textron Specialty Materials SiC fibers such as SCS-6 and SCS-9 with nominal diameters of 143 and 75 μm, respectively. A detailed analysis of the complex microstructure of the SCS-6 fiber is given elsewhere (ref. 7). The CVD type of SiC monofilaments have also undergone diameter reduction as well as improvements in creep resistance, fracture strength, and high temperature properties (ref. 8).

For the past decade, most of the SiC/RBSN composite studies have concentrated on using the latter kind of fiber, i.e. the SCS-6 monofilament (refs. 2 and 3). This limits composite applications since structural components, such as engine nozzles or combustor liners, require more complex reinforcement geometries in which the fibers undergo weaving operations and must accommodate small bend radii. Other disadvantages of using the large diameter monofilament are: (a) low fiber volume fraction (typically below 30 vol%) in the fabrication of SiC/RBSN composites, and (b) limited microcrack-bridging mechanisms. On the other hand, the small-diameter, flexible SiC fiber tow, which does not break when bent in a small radius, offers versatile fabrication capability.

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Also, using multi-filament tows in RBSN composite fabrication should permit higher fiber packing. This should in turn enhance the strength and the toughness of the composite (refs. 9 and 10).

Using small-diameter SiC fiber tow for fabrication of SiC/RBSN composites is feasible only when infiltration of silicon slurry within the tows can be accomplished (ref. 11). The use of sub-micron silicon powders and low slurry viscosity are the criteria necessary to ensure slurry infiltration into the interstices of the fibers. The purpose of this paper is to describe the feasibility demonstration of a fiber coating apparatus and coating procedure which provides continuous slurry infiltration coating of tows. Such coated tows are now employed to fabricate SiC/RBSN composites by heat-treatment to a temperature of 1200 °C or above in a nitrogen environment (ref. 12). Since the coating procedure involves several stages in sequence, the quality of the overall coated tows depends heavily on the processing conditions at each stage and on the processing sequence. The stages used in the coating device are described. Furthermore, the primary factors that influence the coating thickness on the fibers as well as the overall diameter of the coated tows are examined. No optimization for the coating process is being considered in these preliminary feasibility studies because there are too many parameters and/or unknown variables concealed in the process. Finally, the effect of the fiber coating process on the tensile strength of Nicalon fiber is examined.

EXPERIMENTAL

Process Description

The coating device used for coating the fiber tows involves a process which includes several stages. The process starts sequentially when the as-received tows are drawn from the delivery spool and subsequently passed onto the pick-up spool. Figure 1 is a schematic diagram for the coating device that processes the tows continuously. In general, the coating process sequence includes sizing removal, drying, spreading, dip-coating, drying, and die consolidation to infiltrate silicon-filled polymer materials. A continuous coated fiber precursor tow is produced for subsequent fabrication.

Sizing removal is accomplished first by passing the as-received tows into a few cylinders which contain a 50/50 vol% isopropyl alcohol-water solution. The cylinders are placed inside a heated ultrasonic cleaner. The speed of the tow is controlled by D.C. motors (Hurst Motors) and speed controllers (Minarik Electric). The residence time of the tows can be predetermined and monitored. The effects of both the bath residence time and temperature on sizing removal will be discussed in a later section. The wet tows are then passed through a drying furnace before going through the spreading device. Inside the spreading device, air is forced across the passing tows and creates vibrations and some spreading. In addition, electrostatic charges are placed on the moving tows through contact with a static charge generator so as to induce repulsion among the fibers and thus enhance the spreading of the tows. The spread tows are then plunged into the coating device. After the tows are dip-coated with the silicon-filled polymer slurry, they pass through the drying furnace. Finally, as the tows emerge from the furnace, a circular die is used to consolidate the coated tows. Since the rollers and the die that are used in the process are made of Teflon, minimum adhesion and contamination is ensured.

Sizing Removal

The effects of ultrasonic bath residence time and temperature on removal of the M-sizing from the Nicalon tows were studied using an ultrasonic cleaner (Cole-Parmer) operated at a frequency of 40 kHz. Elevated temperatures between 30 to 60°C were implemented using a heater. The amount of residual sizing, after ultrasonic cleaning at a particular temperature and for a given residence time, was determined by the measurement of weight loss using a thermogravimetric balance (Perkin Elmer, TGA 7 series). Since the total wt% of sizing in the as-received tow is about 2 percent, the fraction of the sizing which remains after cleaning is usually very small, on the order of a few tenths of 1 percent. In order to accurately determine the quantity of sizing

remaining using a thermogravimetric balance, a baseline run is incorporated to eliminate any instability and/or buoyancy effects. Duplicate runs (not shown) using the same cleaning procedure indicate a precision of 0.1 wt%.

Slurry Formulation and Characterization

Typical materials used in the process development studies include: (a) Ceramic grade Nicalon fiber tow with M-sizing (polyvinylacetate) in 1800 denier. The tows have nearly 500 filaments with an average diameter of 15 μm. (b) Silicon-filled polymer slurry with particles of 0.3 μm mean diameter were suspended in isopropyl alcohol containing a dissolved polymer binder (Luviskol VA). Various silicon filled polymer solutions were formulated with different concentrations and with different solution properties. The properties of surface tension, viscosity, and density of each concentration were measured at 25 °C by using the drop-weight method, HAAKE viscometer, and hydrometer, respectively.

Coating Parameters

Since the overall diameter of the coated tow, as well as the individual coating thickness obtained from the coating experiments, depends on several parameters used in coating, it would be helpful to distinguish the influence of these parameters on the coating. In general, there are three groups of parameters used for the coating experiments. They are the processing parameters, material parameters, and the geometrical parameters. The processing parameters include the furnace operating temperature for drying the wet coated tows. In addition, the variables used in the dynamic operation of dip-coating, such as the slurry viscosity, surface tension, and tow withdrawal speed, are also included in the processing parameters. On the other hand, material parameters that are directly related to the starting materials, such as the concentration of the coating slurry and the volume fraction of silicon in the dried coating, are grouped separately. Finally, the geometrical parameters, which are the circular die diameter and the die length/diameter ratio, that affect the consolidation of the coated tows, are classified as the third group. All of these parameters can be related, independently or synergistically, in affecting the final coating. The purpose of these preliminary studies was to identify the most influential of these parameters on the coating properties. Then, in later studies, one can optimize the coating parameters to achieve the ultimate properties for the coated tows. The values for the parameters used in this study are listed in Table I.

Coating Theory

The most useful equation used in predicting the liquid coating thickness on a curvilinear surface is the small-wire theory (ref. 13) which is described by equation 1:

$$t_p = \frac{c}{d_p} \times \left(\frac{1.33 Ca^{\frac{2}{3}}}{1 - 1.33 Ca^{\frac{2}{3}}} \right) r \qquad (1)$$

The theory expresses, t , the wet coating thickness as dependent on the radius of the fiber, r. Thickness is also proportional to the capillary number, $Ca = \eta U/\sigma$, where the capillary number is related to the slurry viscosity, η, withdrawal speed, U, and the surface tension of the slurry, σ. One must, of course, assume that the fiber surface is wettable by the coating liquid. However, the most relevant variable is the dried coating thickness, t_p. Eq.(1) shows a term, c/d_p where c is defined as the concentration of particulates in the slurry in g/cm^3, and d_p is the density of dried coating in g/cm^3. Based on the processing and the material parameters used during coating, the solid coating thickness can be estimated using the modified small-wire theory. Recently, in the dip-coating

experiments using monofilament, the coating thicknesses were shown to be in agreement with the predictions (ref. 14).

Coated Tows

The consolidated coated tows that emerged from the die in the drying furnace were cut with scissors into several specimens. The cut specimens were further heat treated in an oven at 100 °C for 15 min to ensure complete dryness. More than 7 specimens of length >25 mm and diameter <1mm were optically examined. These specimens were mounted using fluorescent (solvent yellow 43) resin. The vacuum infiltration technique was used to enhance resin infiltration into the tows. Possible end effects due to cutting and handling were eliminated by taking quarter inch cuts from both sides of the mounted sample. After the samples were polished properly, their morphologies were examined for both global appearance and distribution of fiber.

EXPERIMENTAL RESULTS

Sizing Removal

The success of continuously removing the sizing from the tows is a prerequisite to passing the tows to the next stage. In general, there are two approaches. Thermal decomposition of the tows through an open furnace usually serves the purpose. However, the effects of using higher temperatures (400 to 600 °C) to decompose the sizing in the presence of an oxidizing environment (usually air) may cause detrimental artifacts on the surface of the fibers (ref. 15). These artifacts may become flaws that will affect the fiber strength properties. Furthermore, any debris that remains after using thermal decomposition may require an additional cleaning step. Another alternative is wet cleaning (ref. 16). This route involves using different liquids to accomplish the sizing removal. Oxidizing and/or etching liquids are frequently used in either a single or multi-step operation. However, an easier approach is to use a 50/50 vol% solution of distilled water and isopropyl alcohol at elevated temperatures (35 to 55 °C). The M-sizing, which is polyvinylacetate, is normally insoluble in a static isopropyl alcohol-water solution at elevated temperatures. However, using an ultrasonic device, solubility may proceed with time at elevated temperatures.

The TGA curves in figure 2(a) show the effect of ultrasonic residence time on sizing removal using a constant bath temperature of 50 °C for 3 and 12 min cleaning times. Likewise, the effect of ultrasonic bath temperature on sizing removal using a constant cleaning time of 12 min are shown in figure 2(b). Figures 3(a) and (b) summarize the results obtained from the studies on the effects of ultrasonic bath time and temperature, respectively, on the sizing removal. A typical 90 wt% removal or better can be accomplished using a cleaning procedure of 50 °C for 12 min. Figure 4 shows SEM micrographs of the surfaces of the as-received fibers and fibers subjected to a cleaning procedure of 50 °C for 12 min. It can be seen that it is difficult to infiltrate the tangled as-received fibers while infiltration is feasible when the sizing is almost completely removed.

Coated Tows

The physical properties of the various slurry concentrations used in the coating process were measured by using several instruments. The measurements of surface tension, viscosity, and density for each slurry used are listed in Table I.

The mounted samples of the coated tows were first examined using optical microscopy at 128 X magnification. With the fluorescent mounting resin, one can distinguish the silicon-filled polymer matrix from the mounting resin. From these low magnification micrographs, inter-fiber voids are obvious in all of the samples. The cross-sectional

shape of the overall coated tows varied from specimen to specimen, but the total area within the tow perimeter was integrated and used to calculate the average circular diameter for a given sample. Table I lists the overall mean tow diameter together with the coating conditions for each sample. Statistical deviations from the mean as well as the number of specimens used in reporting the average were included. Duplicate runs were used and included in the table.

The data suggest that, when other coating parameters are held constant, the coated diameter increases as the die (drying) temperature is increased. At low die temperature, 110 °C, the overall tow diameter is smaller than for those generated at higher temperatures. At a die temperature of 170 °C, the overall diameter was found to be independent of the existence of the die, the L/D ratio, withdrawal speed, and slurry viscosity. Furthermore, at a higher die temperature, 190 °C, the coated tow broke apart. Figures 5 to 7 show a set of 4 specimens of polished cross-section micrographs of the coated Nicalon tows processed at die (drying) temperatures of 110, 170, and 190 °C, respectively.

Optical micrographs at a magnification of 1500X were taken to examine the coating thickness on individual coated fibers. The majority of the fibers were distributed randomly within the silicon-filled polymer material. Voids are present in all of the samples studied so far. Also, cracks within the inter-fiber coating material were more numerous for those samples produced at higher temperatures. Only rarely was an individual fiber found to be isolated from the rest of the fibers. For those fibers that were located in the vicinity of the voids, only the coating thickness on the fiber surfaces exposed to voids were examined. The average coating thickness was determined by examining several specimens from the same run (C16-1) and was on the order of 0.5 μm. Based on the coating parameters used, the coating thickness prediction from equation 1 is about 0.3 μm.

Finally, the effects of the fiber coating process on the tensile strength of the fibers were studied. The results (fig. 8) showed that the passing of the tows through the device alone (without any coating slurries) did not affect tensile strength properties at room temperature. A total of 20 samples with a gauge length of 25 mm were used for both the control and the test runs.

DISCUSSION

Although the use of several stages in the fiber coating device to coat a tow seems complicated, it may be simpler than other matrix infiltration approaches. That is, slurry infiltration (refs. 17 and 18) is just one of many methods that can be used to form continuous fiber composite precursors. Other approaches include using electrostatic deposition (refs. 19 and 20), electrophoretic deposition (ref. 21), melt infiltration (ref. 22), and chemical vapor infiltration (ref. 23). These approaches are dependent on many difficult to monitor variables. The dip-coating procedure that is used here may be amenable to easier control because the dip-coating variables and/or parameters are easily predetermined. In addition, the inter-fiber matrix material thicknesses can be monitored by using the modified small wire theory.

In this study, the only pertinent variable that dramatically influences the overall diameter appears to be the drying temperature. From the experimental results, the measured overall diameter of the coated tow is increased as the drying temperature used in the process is increased. At low drying temperature, 110 °C, the overall diameter of the coated tows are more likely to depend on coating parameters, such as the slurry viscosity, concentration, and withdrawal speed. The low drying temperature diameter may even be related to the geometrical parameters to a certain extent, but further investigation is needed to confirm that. However, as temperature is increased while holding other parameters constant, the overall diameter of the coated tows does not depend on the coating parameters as in the case of using low temperature drying. This may suggest that the drying effect, which is temperature dependent, plays a role in controlling the global morphology of the coated tows.

Finally, the correlation between the vapor pressure of the isopropyl alcohol (IPA) evaporated at temperatures above its boiling point (82 °C) should shed light on the global morphology of the coated tows. Using either the Watson or the Chen equation (ref. 24), the heat of vaporization of IPA at three different temperatures (110, 170, and 190 °C) used in processing can be calculated. These values for heat of vaporization, when coupled with the Clausius-Clapeyron equation for an ideal gas behavior, enable the partial pressure of IPA to be calculated at those temperatures. Table II shows the vapor pressure of IPA normalized to the vapor pressure at its boiling point. At 110 °C, the ratio is less than ten, however, at 170 °C, the ratio is increased by 2 orders of magnitude. Further increasing the temperature to 190 °C shows the ratio of the pressures can approach one thousand. Based on the assumption that higher pressure ratios can enhance the degree of separation of fibers during evaporation, this information suggests why the morphology of the coated tows can be transformed into larger overall diameters when the drying temperature is raised.

CONCLUSIONS

A fiber coating device was built and a coating procedure has been developed to coat Nicalon fiber tows continuously with the silicon-filled polymer slurry. The stages used in the coating process are described and the overall coating procedure is examined. In general, the fiber coating device can be used to coat not only Nicalon fiber tows, but also other small-diameter fiber tows to give continuous matrix-coated fiber tows as precursors for composite fabrication. Its use for making small-diameter SiC fiber-reinforced RBSN composites has been demonstrated (ref. 25).

Among the parameters that were described in conjunction with the coating process, the temperature used to dry and to consolidate the wet tows is the most important factor in controlling the global morphology of the tows. Since the vapor pressure of the solvent within the tows is highly temperature dependent, its evaporation is apparently related to the morphology and overall diameter of the coated tows. Once the temperature profile of the wet coated tow is simulated for the drying process, it can be used with other parameters to minimize the inter-fiber voids within the coated tows as well as to optimize the overall quality of the coating.

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TABLE I. —THE EFFECTS OF COATING PARAMETERS ON OVERALL MEAN DIAMETER OF THE COATED NICALON TOWS

Material parameters:			Processing †† parameters:			Geometrical parameters:		Experimental results:	
Sample ID	Vol fr Si in dried film	Conc. of solids in slurry, ς gm/cm^3	Slurry viscosity, η cP	Withdrawal speed, U cm/s	Die temp., T °C	Die diameter, D mm (mils)	Die L/D ratio	†Overall mean tow diameter mm (mils)	Deviation from the mean +/- mm (#)*
C15–1									
110–4–44	0.67	0.53	64	0.5	110	0.71 (28)	4	0.51 (20)	0.03 (7)
170–4–44	0.67	0.53	64	0.5	170	0.71 (28)	4	0.65 (26)	0.03 (7)
170–4–56	0.67	0.53	64	1.3	190	0.71 (28)	4	0.80 (31)	0.05 (7)
170–8–44	0.67	0.53	64	0.5	170	0.71 (28)	8	0.69 (27)	0.02 (7)
170–8–56	0.67	0.53	64	1.3	170	0.71 (28)	8	0.70 (28)	0.04 (7)
170–0–56	0.67	0.53	64	1.3	170	N/A	N/A	0.69 (27)	0.04 (7)
170–0–50	0.67	0.53	64	1.0	170	N/A	N/A	0.67 (26)	0.04 (7)
C16–1									
110–8–56	0.72	0.57	46	1.3	110	0.71 (28)	8	0.55 (22)	0.03 (14)
C16–2									
110–56–28	0.72	0.49	40	1.3	110	0.71 (28)	8	0.59 (23)	0.03 (7)
170–56–0	0.72	0.49	40	1.3	170	N/A	N/A	0.63 (25)	0.04 (7)
190–56–0	0.72	0.49	40	1.3	190	N/A	N/A	0.82 (32)	0.04 (7)

N/A — Without die
(#)* — Number of samples used
† Measured diameter based on circular cross-section
†† —Slurry surface tension ≅ 21.7 dyne/cm

TABLE II.–THE EFFECT OF VAPOR PRESSURE OF THE SOLVENT, IPA, NORMALIZED AT THE BOILING POINT (bp) AT THREE DIFFERENT TEMPERATURES

Temperature	Chen's equation, heat of vaporization	Watson's equation, heat of vaporization	Chen's equation, p(T)/p(bp) ratio	Watson's equation, p(T)/p(bp) ratio
(°C)	(KJ/mol)	(KJ/mol)		
110	36.9	35.9	7	6
170	28.8	28.1	281	243
190	25.1	24.5	1034	861

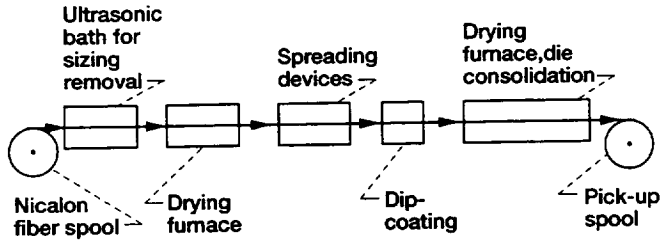


Figure 1.—Schematic Diagram Showing the Fiber Coating

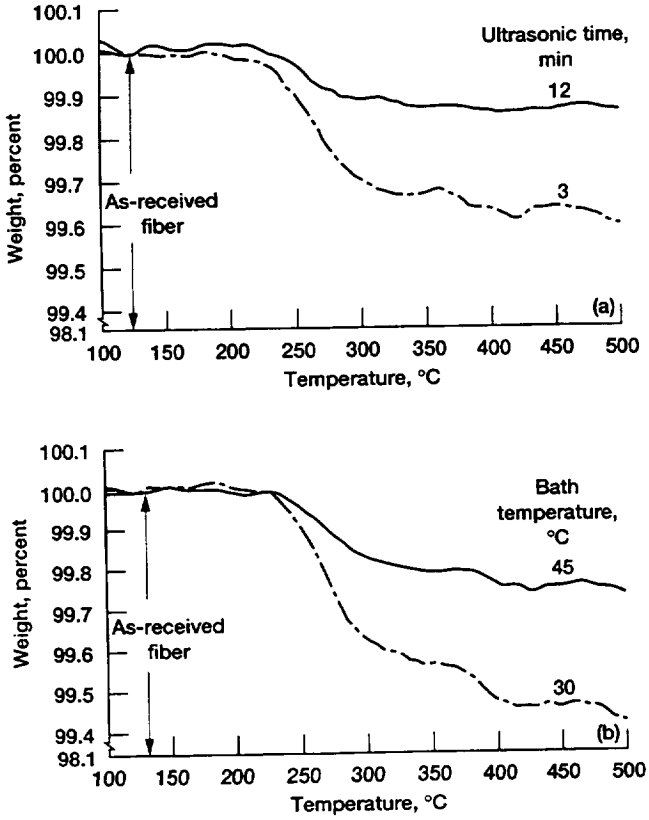


Figure 2.—Effect of ultrasonic bath residence time on sizing removal from Nicalon fiber tow. (a) Bath temperature:50 °C. (b) Residence time:12 min..

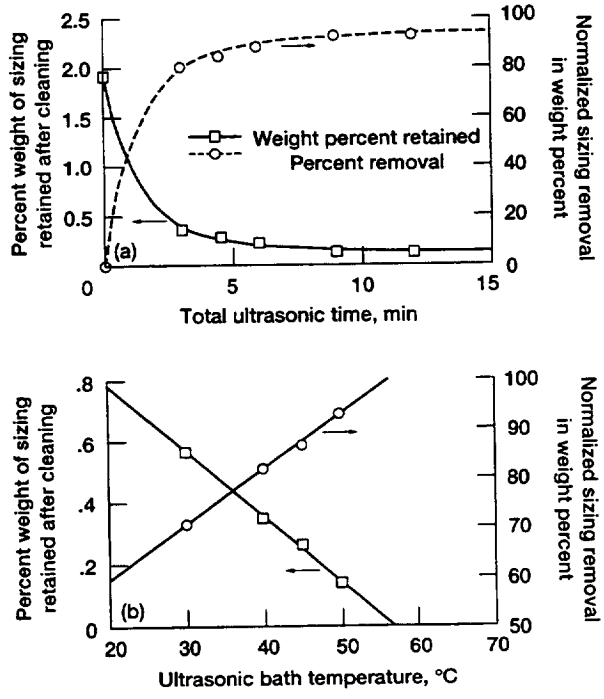
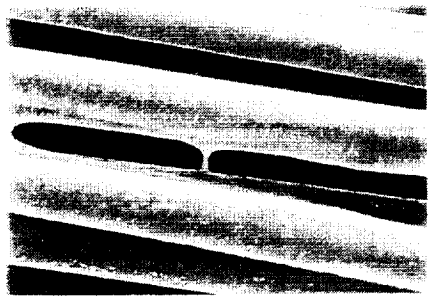
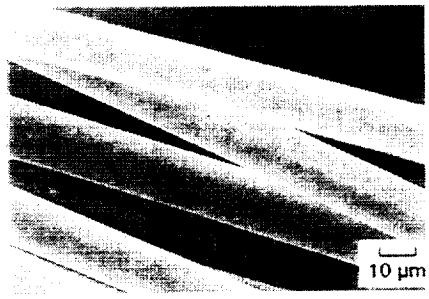


Figure 3.—Effect of ultrasonic time on sizing removal from Nicalon fiber tow. (a) 50 °C bath temperature and 50/50 vol. % isopropanol water solution. (b) Residence time: 12 min. and 50/50 vol. isopropanol water solution.



(a)



(b)

Figure 4.—SEM micrographs showing surface morphology of Nicalon fiber. (a) As received condition. (b) Ultrasonic at 50 °C for 12 min.

Figure 5.—Optica micrographs Nicalon coated tows processed at a die temperature of 110 °C.

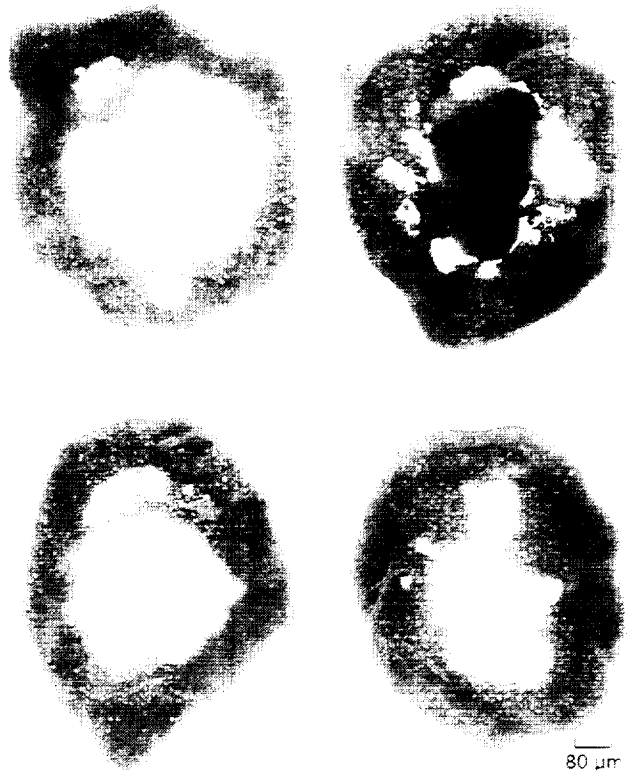


Figure 6.—Optica micrographs Nicalon coated tows processed at a die temperature of 170 °C.

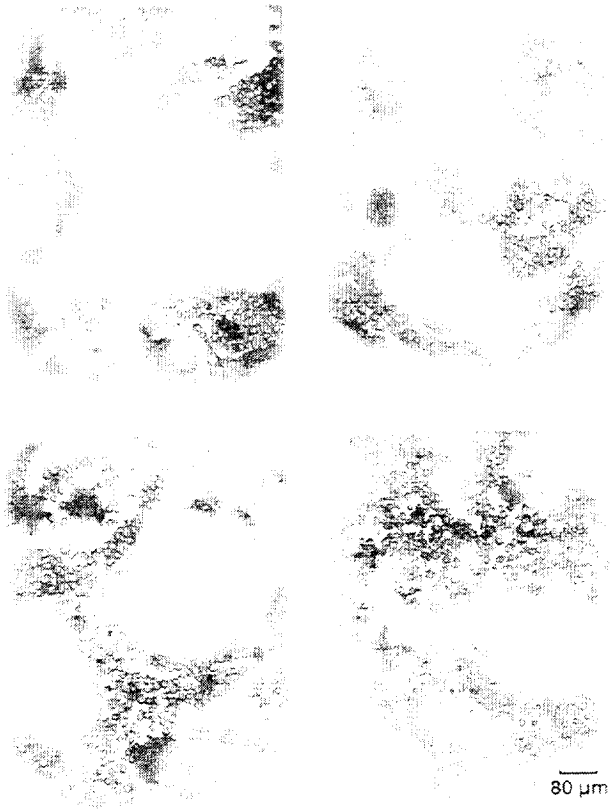


Figure 7.—Optica micrographs Nicalon coated tows processed at a die temperature of 190 °C.

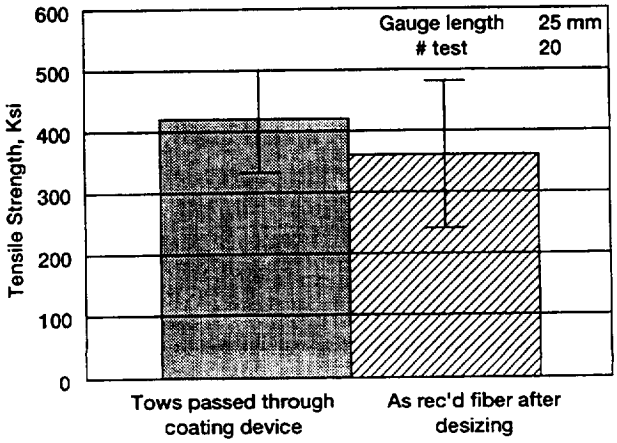


Figure 8.—Slurry coating does not degrade Nicalon fiber strength.

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A process is described for coating continuous small-diameter fiber tows with silicon-filled polymer slurries. This can enable fabrication of complex shaped ceramic matrix composites such as those based on reaction-bonded-silicon-nitride matrices. The coating process sequence includes sizing removal, drying, spreading, dip-coating, drying, and die consolidation to infiltrate a silicon-filled polymer slurry into interstices of the fiber tows. Sizing removal from the as-received fibers was accomplished by using an ultrasonic cleaner with water-isopropyl alcohol solutions at elevated temperatures (30 to 60 °C). Measurements of surface tension, viscosity, and density of various slurry concentrations are listed. The coating thickness applied on the fiber surface can be correlated with the modified small wire theory. Optical microscopy of cross-sections of coated samples showed the degree of infiltration achieved. The overall diameters of the coated tows depended primarily on the drying (die) temperatures used in the drying furnace, which in turn appears related to the vapor pressure of the solvent evaporating at those temperatures. Finally, the effect of the coating process on the tensile strength of Nicalon fiber is also examined.

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